CATALYTIC COAL GASIFICATION IN A DRAFT-TUBE SPOUTED BED BY USING CERAMIC PARTICLES AS A THERMAL MEDIUM

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INTRODUCTION

Steam gasification of coal is thought to be an important future process for production of hydrogen, which is a nonpolluting energy. We have proposed a draft-tube spouted bed type reactor as a catalytic coal gasifier to produce hydrogen.(1) Some minor modifications have been applied to concept of the gasifier. The schematic diagram of the gasifier is illustrated in Fig. 1. In this apparatus, coal is fed to the devolatilization zone in the annulus section. After devolatilization, the char falls into next zone and is gasified by steam supplied from bottom conical section. The heat of gasification is supplied mainly from circulating ceramic particles, which obtains thermal energy from burning gas within the draft tube and returns to the annulus top.

In previous studies (2,3), cold model experiments were carried out by using an acrylic cylindrical apparatus and by using an acrylic semi-cylindrical one. From the results, flow regimes in a spouted bed with a draft tube were clarified, and several correlations concerning the annulus gas velocity and particles circulation rate were proposed for each flow regime. In next step (4), a cold model apparatus, which has the same dimensions as those of the hot model apparatus used in this study, was constructed and was employed to obtain basic data for the hot model operation.

In this paper, we present the results obtained by operating a hot model apparatus of a spouted bed with a draft tube at 1073 K. The hot model unit was heated by electric furnace in the present study, while the unit was designed in consideration of mass and heat balance.

EXPERIMENTAL

Catalytic steam gasification was carried out in a spouted bed with a draft tube shown in Fig. 2. Figure 3 is the photograph of the unit. The bed was heated by four pieces of electric furnace, which surrounded the bed as shown in Fig. 4. The unit includes a stainless steel spouted bed (150 mm I.D.) with a stainless steel coaxial draft tube (10.9 mm I.D.). The draft tube spacing above the gas inlet nozzle (10.9 mm I.D.) was 3 mm. Air was introduced from the nozzle to spout and recirculate solid particles. The bottom of the column was a conical section with a cone angle of $\pi/3$ rad. Steam was introduced through 40 holes (2.0 mm in diameter) located in the conical section. Figure 5 shows the top view of the bottom conical section before assembly. Steam inlet holes (20x2) are found to be arranged concentrically on the conical section. Draft tube was assembled as shown in Fig. 6.

Table 1 shows the physical properties of ceramic particles and coal particles before devolatilization. The ceramic particles (thermal medium) were Nikkato YTZ ball (95% ZrO_2 and 5% Y_2O_3). The char particles were obtained by the following treatment of a bituminous coal of Alaska (Table 2). A sieved fraction of 14 to 20 mesh (average diameter = 1015 μm, Table 1) coal was impregnated with potassium carbonate (gasification catalyst) aqueous solution at room temperature for 3 h. The slurry of the coal was dried up by a rotary evaporator at 323 K. The coal was devolatilized by a fluidized bed (15.5 cm I.D.) at 1123 K for 7 min. The charged amounts of coal were 1.15 kg and 2.08 kg, respectively. Nitrogen (superficial gas velocity at 1123 K = 1.0 cm/s) was used as the fluidizing gas. The catalyst content was 3.4 meq-K/g-char. The minimum fluidizing velocity of the char was 0.79 cm/s at 1073 K.

Table 1 Characteristics of solid particles

solid particles	average diameter [μm]	particle density [kg/m^3]	bulk density [kg/m^3]	terminal velocity [m/s]	minimum fluidizing velocity [m/s]
ceramic	500	6.0×10^3	3.7×10^3	7.1	0.48
coal	1015	1.2×10^3	7.2×10^2	4.9	0.41

Terminal velocity was calculated. Others were measured. Terminal velocity and minimum fluidizing velocity are the values at 298 K.

Table 2 Composition of bituminous coal

ash [%]	VM [%]	FC [%]	fuel ratio	elementary analysis [%] S	C	H	O	N
8.7	42.2	35.0	0.83	0.02	62.9	4.5	21.3	0.9

mineral [%]

Al_2O_3	SiO_2	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	SO_3	P_2O_5	MnO	V_2O_5
14.9	42.3	5.98	23,3	3.42	0.58	1.16	0.85	4.95	0.14	0.24	0.01

The ceramic particles (4.5 kg) were packed into the annulus section. The air was sent through an air filter, an air-oil separator, and an orifice meter before entering the bottom of the bed. The air feed rate was varied to maintain the linear gas velocity within the draft tube as constant (ca. 30 m/s) as possible. Steam was supplied from a vaporizer. The feed rate was controlled by an electric power supply. The particles flowed down through the draft tube spacing above conical section, and were conveyed vertically in the draft tube. At the top, the particles were separated from the carrier gas by a cyclone and returns to the annulus. The temperature of the bed was elevated to 1073 K under spouting by air. As the temperature reached 1073 K, continuous char-feed (0.16 g/s) was started by a rotary feeder. The gasification operation was carried out for 30 min under the conditions summarized in Table 3.

Table 3 Experimental conditions

Gasification temperature	980-1100 K
Packed amounts of ceramic particles	4.5 kg
Circulating rate of ceramic particles	$5x10^{-3}$ kg/s
Feed rate of char	$1.6x10^{-4}$ kg/s
Feed rate of air	2.8 N-m^3/h
Feed rate of steam	0.99 N-m^3/h
Tube-cone clearance	$3x10^{-3}$ m

RESULTS AND DISCUSSION

Figure 7 shows the results of temperature and gas composition. The annulus bed temperature decreased with time, while the external temperature (between upper and lower jacket heater) was maintained at almost constant temperature (990 K). This means poor thermal conduction within the moving bed of the annulus. The gaseous products were hydrogen, carbon monoxide, carbon dioxide and methane as shown in Fig. 7. Hydrogen and carbon monoxide were major products. Since the ratio is unity approximately, the following reaction is thought to be major.

$$C(char) + H_2O \rightarrow CO + H_2$$

The molar rate of $(CO+CO_2+CH_4)$ produced was about $7x10^{-3}$ mol/s during the gasification operation. About half of the product gas was exhausted from the outlet of the annulus. Another half was exhausted from the cyclone. Carbon feed rate was $8.5x10^{-3}$ mol/s, which was calculated from the char feed rate, 0.16 g/s, and carbon content of the char, 64wt%. From these values, we obtained the carbon conversion, ca. 80 %.

The experimental data shows that the draft-tube spouted bed type gasifier is superior to product hydrogen, while poor thermal conduction is a significant problem. Another problem is the bypassing of the product gas to the cyclone.

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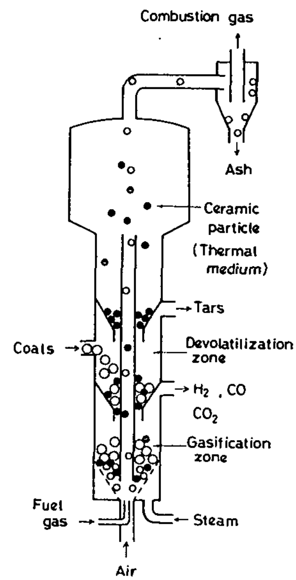
Figure 1. Schematic diagram of gasifier.



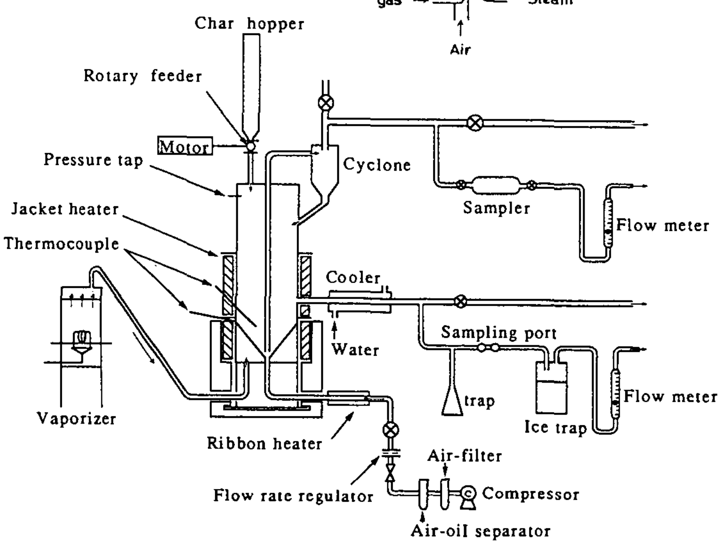


Figure 2. Schematic diagram of hot model unit.

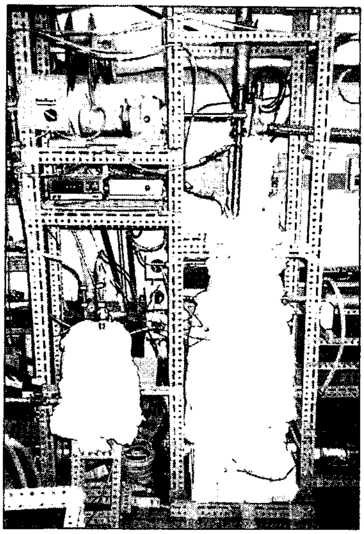


Figure 3. Photograph of hot model unit.

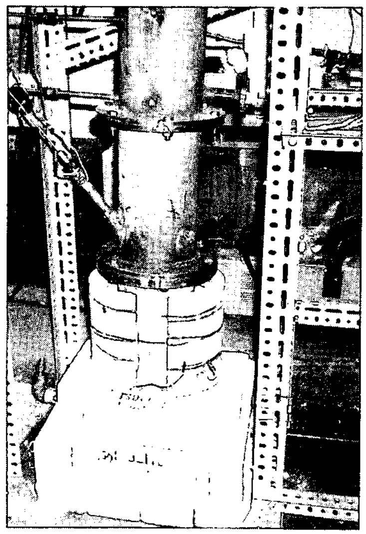


Figure 4. Photograph of hot model unit (under construction).

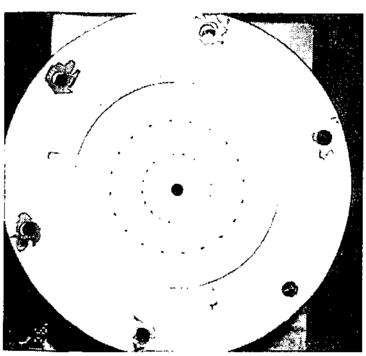


Figure 5. Photograph of bottom conical section.

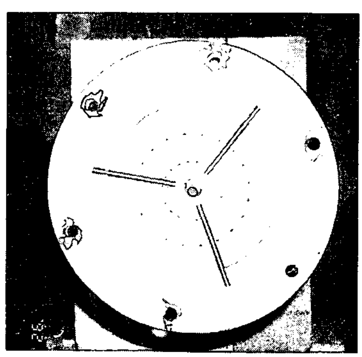


Figure 6. Photograph of bottom conical section with draft tube.

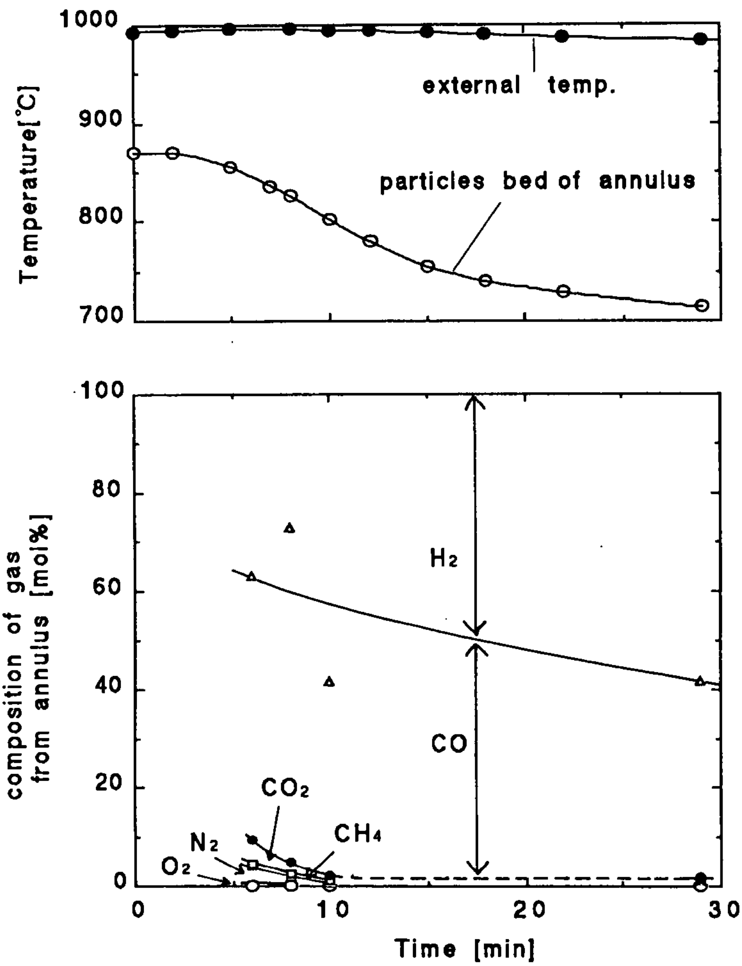


Figure 7. Temperature and gas composition.